

Lawrence Livermore National Laboratory

ENVIRONMENTAL SCIENCES DIVISION

H71

October 18, 1989

Mr. Harry U. Brown, L-505 Department of Energy Nevada Operations Office P. O. Box 98518 Las Vegas, NV 89193-8518

Dear Harry:

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Enclosed are my thoughts on this whole idea of using the Johnston Atoll TRU Clean-up Plant to try to reduce the Pu concentration in the surface soil at Rongelap Island. My major concern is the extreme environmental impact to Rongelap Island resulting from such an operation for a truly insignificant reduction in the total estimated dose received from living on Rongelap. Plutonium and Americium contribute less than 1% of the estimated 50-y integral effective dose equivalent and yet you would have to literally denude the island to do the project.

In addition to the extreme environmental insult, I think there are technical reasons why the system may not work at Rongelap as it does at Johnston Atoll.

My feeling is that if a serious effort is to be made to reduce the dose equivalent at Rongelap Atoll below the currently estimated low level of 40 rem/y and 0.9 rem is 50-y, it should be directed toward 137Cs and the terrestrial food chain for which we have an effective countermeasure.

Best regards,

William L. Robison Terrestrial and Atmospheric Sciences

WLR:lj

Encl.

HARRY BROWN'S Files, NV

COMMENTS ON THE USE OF THE JOHNSTON ATOLL "TRU CLEAN" SOIL CLEAN-UP PLANT

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The "Tru Clean" soil decontamination plant at Johnston Atoll was designed to accomplish the very specific task of separating plutonium oxide (PuO), or PuO particles attached to soil particles (CaCO3), from the bulk surface soil. The contaminated soil is passed over a detection system which measures the amount of 241Am in the soil to determine if the soil can be considered "clean" or whether the soil should be processed to remove transuranic radionuclides. The detection limit for the system, or in other words soil that is considered "clean," is 500 Bq/Kg (13.5 pci/g) for Pu+Am.

There are several reasons why this system probably should not be considered for use at Rongelap Atoll. They are:

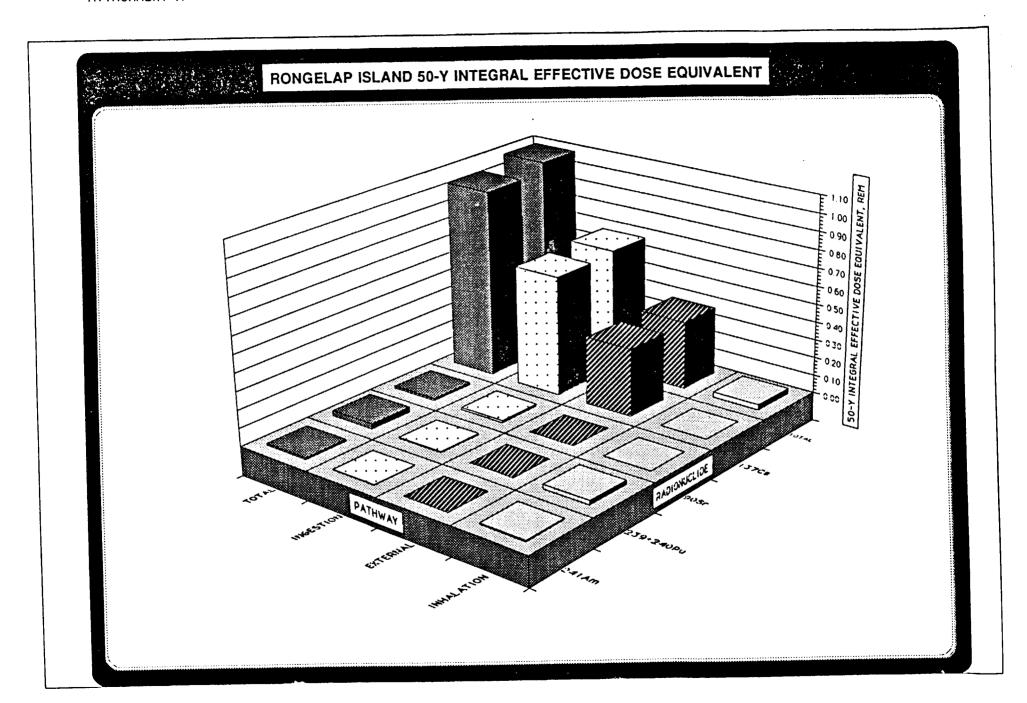
1. Environmental Impact

To attempt to separate the Pu in the top 15 to 25 cm of soil at Rongelap Island would require removing all of the coconut, breadfruit, <u>Pandanus</u> and lime trees and all other vegetation. The negative impact in the island is tremendous. It seems hard to justify such an insult to the island's ecosystem and the long term revegetation process when the potential dose from Pu and Am at Rongelap Island is so small (see Attachment A and B).

2. <u>Different Form of Pu at Rongelap Atoll</u>

The transuranic radionuclides at Johnston Atoll are primarily in the oxide form (PuO and AmO). This is because the source of the Pu was a device that was destroyed on the launch pad by high explosives and thus did not go nuclear. As a result, the Pu oxide metal, with a density of about 11.5 g/cm^3 , was dispersed into the surface soil at Johnston Atoll. The density of coral soil is between 1.2 and 1.5 g/cm^3 . Consequently, because of the vast difference in density between PuO and coral soil (11.5 g/cm^3 versus 1.5 g/cm^3), the PuO can be separated from the coral soil by gravimetric means.

At Rongelap, on the other hand, the source of Pu is the Bravo event which was a nuclear detonation. The transuranic radionuclides were atomized at the time of detonation and recondensed in or on carbonate soil particles that were engulfed in the fireball at detonation. The larger size carbonate particles in the Bravo cloud settled-out early on and smaller size carbonate particles were carried downwind about 90 miles and deposited on Rongelap Atoll. As



THE RADIOLOGICAL DOSE FROM Pu AT RONGELAP ISLAND

W.L. Robison
C. Sun
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- A. Introduction
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 - 3. Summary-Comparison Environmental and Urine Analysis

RONGELAP ISLAND

INTRODUCTION

The important issue to focus on when plutonium (Pu) and Americium (Am) are present in the environment is the potential radiological dose to people living in that environment; the dose can be directly related to risk and biological effect. There are two basic methods for estimating this dose; one we will refer to as the "environmental method" and the other as the "urine analysis method." Other issues, such as the concentration of Pu in soil, are only relevant insofar as they provide information for the environmental method.

DOSE ESTIMATES

Environmental Method (Lawrence Livermore National Laboratory)

Pu in the soil is of no consequence if it is neither ingested nor inhaled. Thus, when Pu is present in the environment the potential radiological dose must be evaluated for both the inhalation and ingestion pathways in order to estimate the potential biological effect. The radiological dose is dependent on the uptake of Pu by food crops and their subsequent ingestion by people, possible direct consumption of surface soil, and resuspension by wind of surface soil particles in the respirable size range that contain Pu which can be inhaled.

Uptake of Pu by food crops and resuspension of Pu contaminated surface soil are very dependent on environmental variables such as soil composition, soil pH, vegetation ground-cover, height of the vegetation canopy, and suspendability of the surface soil. If data are available for the uptake and resuspension of Pu for a specified environmental system, then these variables are accounted for and a direct and meaningful comparison can be made on the critical issue-the potential dose to people living in a specified environment.

We have analyzed many vegetation samples in the Marshall Islands, including Rongelap Island, to determine the concentration of Pu and Am in food crops. We find that plants have a very, very low uptake of Pu and Am and the consumption of soil is minor, being limited to occasional dust on ones hands. As a consequence, resuspension of plutonium contaminated surface soil, and the subsequent inhalation of Pu contaminated dust particles in the respirable size-range,

is the major <u>potential</u> route of exposure to people in the Marshall Islands as it is in almost any environment.

The resuspension of surface soil varies greatly, however, from one environment to another; resuspension may be very high in one environment and essentially negligible and of no consequence in another. Thus, it is much preferred that data for the concentration of Pu in air be available so that models can be developed relating Pu air concentration to Pu surface soil concentration, thereby eliminating much of the uncertainty in predicting resuspension mechanisms for a specific environment. We also have extensive data on the Pu and Am concentrations in surface soil and air from which we can estimate the amount of Pu and Am which might be inhaled or ingested during residence on Rongelap Island.

The 50-y integral effective dose equivalents for both the ingestion and inhalation pathways are based on the following:

Ingestion

- 1. The average concentration of Pu and Am measured in food products from Rongelap Island.
- 2. The ingestion of local foods based on the diet listed in Table A-1 of the attached Appendix A.
- 3. An assumption that 10 mg per day soil is ingested for every day of a person's life. We think this is conservative in that it overestimates the actual soil consumption of adults over their lifetime.

Inhalation

1. The average Pu concentration in air based on the LLNL resuspension model for Rongelap Island is conservatively <u>estimated</u> to be 190 aCi/m³. This concentration is assumed to be present every day of a person's residence on Rongelap Island and when combined with the average breathing rate of 22 m³/d gives the daily Pu inhalation rate in aCi/d. For comparison, the <u>measured</u>, average background concentration of Pu in air at Bikini Island at Bikini Atoll and Enjebi Island at Enewetak Atoll,

where the Pu concentration in the surface soil is 3 to 4 times higher than at Rongelap Island, is only about 30 to 60 aCi/m³ (1,2). Consequently, the average Pu concentration in air which we use to estimate the dose from inhalation is very conservative and, if anything, will probably overestimate the potential dose to people living on Rongelap Island.

2. The inhalation model as given in references 1 and 3.

The effective committed dose equivalent based on the above data is 75 mrem for Pu plus Am; the 50-y integral dose equivalent is 56 mrem. The relative contribution of Pu and Am and the inhalation and ingestion pathways is listed in Table 1.

To help put the estimated effective committed dose equivalent or the 50-y integral effective dose equivalent from Pu and Am in perspective, we will compare them to the committed effective dose equivalent due to ¹³⁷Cs and ⁹⁰Sr at Rongelap Island, the natural background committed dose equivalent at Rongelap Island and the average natural background committed effective dose equivalent in the United States.

The average effective committed background dose equivalent in the United States is 300 mrem/y (4). Over 50 y this is a total effective committed dose of 15,000 mrem; this is the average dose to a citizen of the U.S. and includes all external exposure and internally deposited radionuclides. The comparable dose at Rongelap Island for all radionuclides plus natural background is 2,200 mrem of which only 75 mrem is due to Pu + Am. The results are listed in Table 2. Based on our conservative estimates of the intake of Pu and Am by ingestion and inhalation, the estimated effective committed dose equivalent of 75 mrem due to Pu and Am at Rongelap Island is 1/200 of the average U.S. background dose. Plutonium and Am account for only 7% of the estimated committed dose equivalent due to man-made radionuclides at the island; ¹³⁷Cs accounts for about 90%.

The same conclusion, that Pu and Am at Rongelap contribute very minor radiation doses, can be reached by calculating an Annual Limit of Intake (ALI) for the general public from values listed in ICRP Publication 30 for radiation workers. An ALI for the public can be estimated by assuming that the ALI is a factor of 50 less than that for workers (5000 mrem divided by 50 equals 100 mrem). The results are shown in Table 3 and are converted from annual to daily intakes. The intakes at

Rongelap for inhalation and ingestion are about 65 to 240 times less than one derives from the ICRP recommendations.

Table 1. The effective committed dose equivalent from Pu for 50 y of residence on Rongelap Island.^a

		mrem	
	Inhalation	Ingestion	Total
Pu	34 (28)	12 (6.3)	46 (35)
Am	<u>23</u> (18)	<u>6.0</u> (3.4)	<u>29</u> (21)
Total	57 (46)	18 (9.7)	75 (56)

^a The 50-y integral dose equivalent is given in parentheses.

Table 2. The effective committed dose equivalent from Pu and Am at Rongelap Island and the effective committed background dose equivalent in the United States.^a

	Effective committed dose equivalent, mrema		
Pu + Am dose at Rongelap	75 (56)		
137Cs + 90Sr dose at Rongelap	1,025		
Natural background at Rongelap	1,100		
Total	2,200		
U.S. background (all radionuclides)	15,000		

^a The 50-y integral dose equivalent is given in parentheses.

Urine Analysis Method (Brookhaven National Laboratory)

In this method the Pu concentration in urine is determined by state-of-art fission track analytical (FTA) procedures. The measured Pu concentration is used in conjunction with excretion models for Pu to estimate the dose from Pu remaining in the body.

Table 3. The annual intake of Pu via ingestion and inhalation at Rongelap Island compared with Annual Limit of Intake (ALI) for the public derived from recommendations by the ICRP for radiation workers. Intakes are converted from annual to daily intakes.

	Pu daily intake, pCi/d			
	Rongelap	ICRP (public)a	Ratio ICRP/Rongelap	
Ingestion	0.18	44	244	
Inhalation	0.0046	0.30	65	

a Derived from ALI recommendations by ICRP for radiation workers (ICRP Publication 30, Part 4, 1988).

As of December 1988, over 500 urine samples collected during 1981 to 1984 from the Rongelap people were completed. Although these measurements have met rigorous quality assurance standards for chemical analysis, some inconsistencies still existed in the FTA data which we presented during the Livermore meeting in February 1988.

Now all 67 urine samples of the Rongelap people taken last September 1988 have been analyzed. The results support the thesis that soil contamination in some of the earlier urine samples was giving false information. Because of BNL's careful attention in September to collecting uncontaminated urine samples, which was facilitated by Majatto's low soil concentration of plutonium, we were not surprised to find the statistics of current Rongelap measurement reflect a median value far below the 250 aCi per sample as presented at the Livermore meeting.

Past studies of plutonium concentration in urine samples obtained from the Marshall Islands people indicated levels much higher than those now known to be present. The new sample data are, in part, the result of improved bioassay sample collection and analytical technology. Furthermore, it now appears that earlier "high" plutonium results were very likely due to: (1) naturally occurring polonium-210 inhaled in cigarette smoke and fresh fish and (2) water and soil contamination of the urine samples during collection.

The polonium problem was resolved by the adaption of our FTA method. Regarding soil contamination of the urine sample, the analyses of the September 1988 samples provided the following information:

- From the samples taken in Majatto, all of the plutonium results are below 170 1. aCi (a committed effective dose equivalent 85 mrem, i.e., the total dose to be received over the next 50 years). The median of the distribution is at the background level.
- 2. An interesting observation is that the plutonium concentrations in the Rongelap people's urine samples is similar to that of our BNL individual who was used as our laboratory control up to December 31, 1988.
- 3. The mean Pu concentration in urine is below the FTA detection limit of 80 aCi; the 50-year effective committed dose equivalent based on the detection limit is about 40 mrem. The actual 50-year effective committed dose equivalent is something less than 40 mrem but how much less is unknown because of the detection limit.

SUMMARY

The radiological dose due to Pu in the environment at Rongelap is estimated by two very different methods (Environmental and Urine Analysis) and compared in Table 4.

The estimated effective committed dose equivalent (or the 50-y integral dose equivalent) due to Pu at Rongelap Island are very similar for the two quite independent methods. It is apparent that there is complete agreement between BNL and LLNL on the magnitude of the dose from Pu at Rongelap Island. Consequently, the 40 to 46 mrem effective committed dose equivalent (35 mrem 50-y integral dose equivalent) from Pu is a very small fraction of the total estimated dose at Rongelap Island which in turn is less than 15% of the effective committed background dose of 15,000 mrem or more in the U.S. and other worldwide locations.

Table 4. The average effective committed dose equivalent from Pu at Rongelap Island in mrem.

	Me	Method		
	Environmental (LLNL) Effective committed dose equivalent	<u>Urine Analysis (BNL</u> Effective committed dose equivalent		
Pu	46 (35) mrem	40 mrema		
Am	29 (21) mrem	Assume Am 2/3 of Pu		

a Based on the detection limit. The actual mean dose is something below this number.

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- 3. W.L. Robison, C.L. Conrado, and W.A. Phillips, <u>Enjebi Island Dose Assessment</u>, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-53805 (1987).
- 4. National Council on Radiation Protection and Measurements, Exposure to the Population in the U.S. and Canada from Natural Background Radiation, National Council on Radiation Protection and Measurements, Washington, DC, NCRP-94 (1987).

APPENDIX A

Table 5. Comparison of the predicted and measured body burdens of ¹³⁷Cs for three atolls in the Marshall Islands

Predicted adult body burdens using dose <u>models and various diet options (pCi)</u> <u>LLNL diet model</u> Imports Imports Communi			lose bo	Measured average body burden in 1978 by BNL (pCi) BNL diet ity Community	
Atoll	available Maximum	unavailable	В	Α	Average
Bikini	5.5 5.7 (N	11 (A)	20	45	2.4 (M) ^a
	2.7 (1	F)			1.7 (F) ^b
Rongelap	0.16	0.42	0.46	1.3	0.17 (A) ^c
Utirik	0.04	3 0.098	0.18	0.35	0.053 (A)

^a Male.

^b Female.

c Adult.